# Magnetic-field-dependent plasma composition of a pulsed aluminum arc in an oxygen ambient

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A variety of plasma-based deposition techniques utilize magnetic fields to affect the degree of ionization as well as for focusing and guiding of plasma beams. Here we use time-of-flight charge-to-mass spectrometry to describe the effect of a magnetic field on the plasma composition of a pulsed Al plasma stream in an ambient containing intentionally introduced oxygen as well as for high vacuum conditions typical residual gas. The plasma composition evolution was found to be strongly dependent on the magnetic field strength and can be understood by invoking two electron impact ionization routes: ionization of the intentionally introduced gas as well as ionization of the residual gas. These results are characteristic of plasma-based techniques where magnetic fields are employed in a high-vacuum ambient. In effect, the impurity incorporation during reactive thin-film growth pertains to the present findings. 
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A variety of plasma-based growth and etch techniques utilize magnetic fields to guide and focus ion beams as well as to increase the plasma density of low pressure discharges. Examples of magnetically enhanced growth or etch techniques are: Magnetron sputtering with internal[1] ($B=0.12$ T) and external[2] ($B=0.013$ T) Helmholtz coils, helicon plasma sources[3] ($B=0.2$ T), magnetically filtered arc[4] ($B=0.18$ T), as well as magnetically filtered high current arc[5] ($B=0.046$ to 0.2 T). An indepth discussion on magnetically enhanced physical vapor deposition techniques is available.[6]

Naturally, vacuum based techniques are characterized by the presence of residual gas. Recently, it was shown[7] that ionization of residual gas occurs in the presence of a 0.4 T magnetic field in a Au arc plasma without intentionally introduced gas, 0.22 atomic ratio of the plasma consisting of ionized residual gas. Metal plasma streams generated by cathodic arc sources are widely used for reactive thin-film synthesis[6] as well as for ion implantation,[8] ion immersion implantation,[9] and ion injection into accelerators.[10] An inherent drawback of utilizing cathodic arc sources for thin-film growth is the production and subsequent incorporation of so called macroparticles. The most frequently applied approach to strongly reduce the amount of those is to utilize curved magnetic fields for macroparticle filtering.[11] There are published data available describing the effect of a magnetic field on the plasma composition of reactive, pulsed cathodic arcs.[12] In this work, the presence of ionized reactive gas was reported. However, no attention was paid to the concurrent ionization of the residual gas.[12] This apparent limitation in perspective may be due to the condition that plasma impurities are not important for applications of ion beams where mass selection is used, for example, prior to injection into accelerators. However, if thin-film growth is attempted, plasma impurities that originated from the residual gas[13] can have a profound influence on the composition and structure of films.[14,15] A meaningful investigation of the plasma composition evolution in the presence of magnetic fields must therefore include all plasma constituents.

Here, we report plasma composition data clearly indicating extensive ionization of (i) the residual gas, as well as the (ii) previously reported ionization of intentionally introduced gas in the presence of magnetic fields. The reported data are quantitatively relevant for filtered cathodic arc techniques as well as qualitatively relevant for plasma-based deposition and etch techniques in the presence of magnetic fields since electrons are already trapped at field strengths in the range of several mT. This is because the electron gyration radius for such field strengths is smaller than the discharge dimension for typical electron temperatures ($T_e$) in a low pressure discharge $T_e \sim 2-5$ eV,[16] and the collision frequency is smaller than the gyration frequency.

The plasma composition analysis was carried out using a vacuum arc plasma source with a time-of-flight (TOF) charge-to-mass spectrometer in which an ion beam is formed by the extraction of ions from the vacuum arc plasma source (distance from source to extraction system ~0.1 m). A simplified schematic of the experimental setup is shown in Fig. 1. A 200 ns sample of the beam pulse is selected by a set of annular electrostatic TOF gating plates (distance from extraction system to TOF gate ~0.65 m) and allowed to drift to a magnetically suppressed Faraday cup[17] which is located 1.03 m from the TOF gate. From the measured ion current–time dependence, the plasma composition can be calculated,

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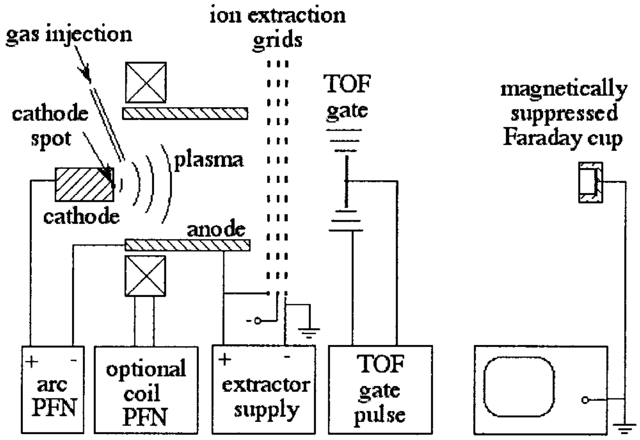


FIG. 1. Schematic of the experimental setup.

since the TOF of the different ionic species in the beam depends on their charge to mass ratio.[17] A typical example of the current versus TOF is shown in Fig. 2. The TOF system has been described in detail elsewhere[17] and has been used extensively to explore the charge state distribution of the ions produced by the vacuum arc plasma.[17–19]

The TOF system was modified in such a way that a magnetic field in the immediate vicinity of the cathode can be induced by a magnetic field coil, powered by an external pulsed power supply. The variation of the magnetic field strength was $\pm 5\%$ over the whole arc pulse, as shown in Fig. 3.

The ion beam extraction is virtually unaffected by the magnetic field, since the magnetic field strength at the extractor location is $<1\%$ of the maximum value in the solenoid center. Hence, the effect of the magnetic field is limited to the plasma at and close to the solenoid. This technique was successfully used in the past to modify the charge state distribution of ions produced by the vacuum arc plasma.[18,19] Data were taken at an oxygen partial pressure of $5 \times 10^{-5}$ Torr (6.7 mPa), the base pressure was $2 \times 10^{-6}$ Torr (0.27 mPa). The magnetic field strength was varied from 0 to 0.36 T. The rectangular 400 A arc current pulse, generated by a 1 Ohm pulse-forming network, had a duration of 250

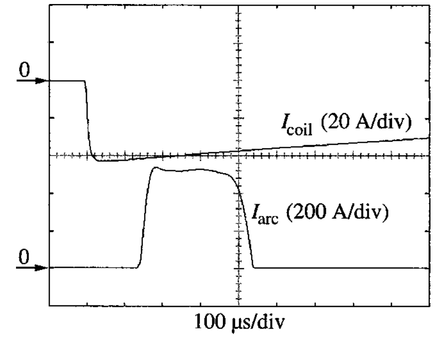


FIG. 3. Magnetic field strength and arc current vs time.

$\mu$s. All data were taken at 150 $\mu$s after the arc was ignited and 50 plasma pulses were averaged for the plasma concentration calculation.

In Fig. 4, the log plasma concentration is shown versus the magnetic field strength. A strong influence of the magnetic field strength on the plasma composition was observed. Without the magnetic field, the plasma consists to 95.1% of ionized Al species; the balance is given by ionized oxygen with 3.0% and 1.9% of ionized hydrogen, which is consistent with previously published values.[20] As the magnetic field strength is increased to 0.22 T, the nonmetal ionized species increase by an order of magnitude ($O = 45.7\%$; $H = 32.6\%$). At larger $B \geqslant 0.22$ T ionization of the nonmetal species have reached saturation. Saturation may be reached as a result of the limited amount of oxygen and water available in the system.

The extensive amount of hydrogen originates from water (from the ambient water and water induced by plasma stimulated desorption from the chamber walls),[7] the main constituent of the residual gas in a high vacuum ambient.[13] It is straightforward to understand that the oxygen ion current is a result of ionized intentionally introduced oxygen and oxygen originating from ionized water. The presence of ionized residual gas is particularly significant in thin-film deposition. It is well known that the absorption probability of an energetic ion into a growing film is often larger than for a low energy neutral (see for example Ref. 21). Previously we have found

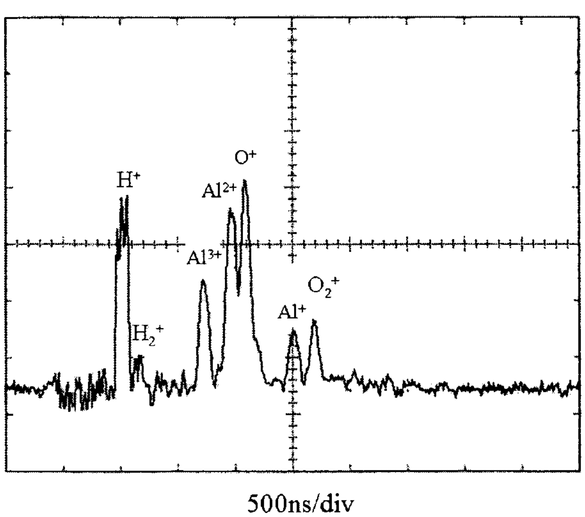


FIG. 2. Ion current vs TOF (500 ns/div) at an oxygen partial pressure of $5 \times 10^{-5}$ Torr and a magnetic field strength of 0.36 T.

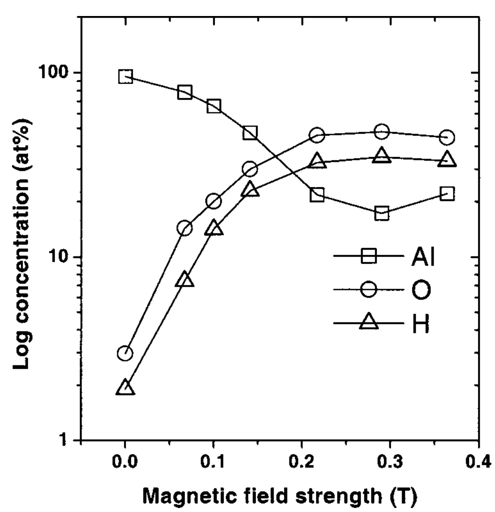


FIG. 4. Plasma composition vs magnetic field strength at an oxygen partial pressure of $5 \times 10^{-5}$ Torr.

that an extensive amount of hydrogen is incorporated in films grown from a similar plasma as analyzed in this work.[14]

It is reasonable to assume that nonionized, residual gas species are present in the vacuum arc plasma in any case, but their ionization depends strongly on the presence of a magnetic field. In the absence of a magnetic field, ionization of the residual gas in the gas phase as well as desorbed species from the walls is comparatively small, because the mean free path of the electrons is large compared to the discharge dimension, and hence electron impact ionization is only minor. The situation is different when a magnetic field is present. Electrons are trapped and gyrate around magnetic field lines; their path length and residence time in the plasma volume of interest is multiplied, and so is the probability of ionizing collisions. This has large implications for all plasma-based deposition and etch techniques in the presence of magnetic fields since our data show that a large fraction of the plasma are impurities stemming from ionization of residual gas. In particular, if thin-film growth is attempted with a filtered cathodic arc, the transport efficiency of the magnetic filter is optimized by the applied magnetic field. Optimized values of the magnetic field strength reported in the literature are for examples 0.18 T (Ref. 22) and 0.1 T (Ref. 5), which would result, according to our data in a hydrogen fraction of the plasma, between 28% and 14%.

The criterion to estimate optimized filter transport was to measure the magnitude of the filtered ion flux. The presented data clearly indicate that a considerable fraction of the filtered ion flux may originate from the ionization of residual and intentionally introduced gas and thus does not exclusively represent the ion flux of cathode material as generally assumed. It is likely that this rather large impurity concentration of the plasma will affect the evolution of the film composition, structure, and, therefore, the film properties. The incorporation of plasma impurities, however, depends on the reactivity of the growing film.

We have shown that for reactive thin-film growth with pulsed plasma systems in a high-vacuum in the presence of magnetic fields, an extensive amount of the condensing flux at the substrate are ionized residual gas species. This may lead to the incorporation of large amounts of impurities in a growing film. These results pertain to all plasma-based techniques in the presence of magnetic fields in residual gas, and in particular for filtered cathodic arc systems.

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